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16. ABSTRACT

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APPLICATION OF MULTIMEDIA EXPOSURE ASSESSMENT TO DRINKING WATER

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Abstract. A potentially important exposure route for humans is the ingestion of chemicals via drinking water. If comprehensive exposure assessments are to be completed for either existing or proposed new chemicals and cost effective control strategies developed, then a quantitative understanding of multimedia transport and fate of specific chemical pollutants must be achieved. Mathematical models provide a powerful framework into which quantitative relationships may be placed to provide guidance in reaching water quality goals. Existing, state-of-the-art media-specific toxic organic transport and fate models for atmospheric (DiDOT), land surface (NPS) and surface water processes (EXAMS) and potable water treatment (WTP) have been linked to demonstrate the technical feasibility of such an approach. Limited application and sensitivity testing of this linked modeling system has shown that the impact of various source loadings and control strategies on drinking water quality can be estimated.

1. Introduction

Ingestion through drinking water is a potentially important route of exposure for humans to toxic substances. If drinking water is derived from groundwater sources, its contamination will usually come from waste disposal sites (e.g., landfills, lagoons). If surface water supplies are used, however, the sources of contamination may be multiple and spatially and temporally variable, and the pollutants may interact as they are transported and transformed among various environmental media prior to their withdrawal in the raw water for a treatment plant. Pollutants can enter surface waters through direct deposition from the atmosphere (dryfall and wetfall), direct wastewater discharges, contaminated runoff from urban or rural watersheds, and contaminated groundwater contribution to stream flows. Primary pollutant sources can include stack emissions, wastewater discharges, and waste disposal sites.

The major or predominant pollutant sources and the routes through which surface waters become contaminated may be obvious in some cases, but in many situations the contamination is indirect and multimedia transport and transformations are involved. Similarly, any control strategy is complicated by the 'residuals shifting' problem. That is, reduction in a release to one media may result in an increased release to another media. For example, reduction in stack emissions may generate more solid waste residuals requiring disposal in a landfill. If cost-effective control strategies are to be developed and

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if comprehensive exposure assessments are to be completed for existing and proposed chemicals, then a quantitative understanding of multimedia transport and fate of pollutants must be achieved. Mathematical models provide a powerful framework into which such quantitative relationships may be placed to provide guidance in reaching water quality goals. This paper describes such a prototype model developed for use in multimedia exposure assessment of organic toxicants.

2. Description of the Modelling Approach

The conceptual basis for exposure assessment modelling in aquatic systems was described by Sanders (1975). His modelling concept identified the environmental transformation processes (physical, chemical, and biological) influencing toxicants in the environment and combined quantitative transformation descriptions with loading rates and transport phenomena to yield projected concentration distributions within selected aquatic environments. This same rationale applied to multimedia assessment has led to the Air, Land, Water Analysis System (ALWAS) developed for the Environmental Protection Agency by A. D. Little, Inc. (Tucker *et al.*, 1981).

The objectives of this ALWAS prototype effort were largely to investigate the feasibility of linking existing media-specific models into a comprehensive system that could evaluate reasonable range of multimedia exposure assessment problems. The approach is based on the premise that processes influencing the pollutant mass or concentration within each environmental media (air, soil, water) can be combined with meteorologic and hydrologic transport mechanisms (atmospheric dispersion, runoff, erosion, stream flow, etc.) to predict selected time and spatial concentration profiles in each media given the appropriate total (by media) loading information as input. A balanced approach is desired that allows consistent levels of approximation or sophistication for each of the various media components. The existing component or media-specific models selected for incorporation into ALWAS according to the above criteria include: (1) the Dispersion and Deposition of Toxics (DiDOT) model (Tucker *et al.*, 1982); (2) the Non-Point Source (NPS) model (Donigian and Crawford, 1976); and (3) the Exposure Analysis Modelling System (EXAMS) (Burns and Cline, 1982). Application of ALWAS to analyze finished drinking water exposure requires addition of a model for the fate of organics in potable water treatment plants. For this final component, the Water Treatment Processes (WTP) Model developed by Hedden (1980) was combined with ALWAS to complete the analysis presented later in this paper.

Mathematical convenience and computational efficiency are also important objectives for a feasibility study of this nature. To maximize investigation of the most important perceived inter-media transfers of organics, an approach was adopted that minimizes feed-back loops between the media-specific models. The approach used avoids the need to run air, land (runoff) and aquatic models simultaneously; rather, each is run independently with outputs from one serving as inputs to the others. Coupling within a watershed defined by aquatic and runoff models is assumed to be from air to surface (land or water) only. Because the residence time of air over a watershed is usually very short when

compared to similar residence times for water- or soil-related processes, the return of organics to the air from land or water (e.g. volatilization from water) is assumed to have a negligable effect on the air mass balance.

3. Description of the Component Processes and Models

Complete descriptions of each process currently known to be pertinent to multimedia assessment is included in the report by Tucker *et al.* (1981) and in the documentation of each component model. Such detail will not be repeated here but specific highlights are given to illustrate how ALWAS evolved and to further elucidate areas of specific strengths and weaknesses.

3.1. Atmospheric Processes

The processes known to be important in estimating atmospheric transport and transformations include dispersion, wet and dry deposition to land or water surfaces, and chemical degradation from direct photolysis, ozone attack, OH radical attack and RO_x radical attack (Tucker *et al.*, 1981). The dispersion phenomena must be applied to individual point sources (specific sources for which stack dimensions are known) and to area sources (a complex of sources to which a uniform source strength is assigned). In the general case, mass transfer from land and water surfaces (e.g., volatilization or wind entrainment) can also occur. Such surface-to-air transport may be important in estimating near-surface ambient air concentrations, but, as stated earlier, are believed to be less likely to greatly influence land or water ambient concentrations (Tucker *et al.*, 1981).

The DiDOT model treats dispersive transport with Gaussian dispersion formulations much like those found in most standard air transport models and is subject to the inherent constraints imposed by such procedures. Atmospheric degradation is modelled by applying pseudo-first order kinetics to the lumped processes of photolysis and O_3, OH^{-1}, and RO_x-reactions. Inherent in this approach is the assumption that the environmental properties (e.g., OH_3, OH^{-1} concentrations) are constant over large representative areas (e.g., rural, urban) thus reducing the fundamental second-order processes to pseudo-first order approximations. Wet and dry deposition processes in ALWAS are formulated somewhat differently than those in standard air quality models. Dry deposition is estimated by accounting for both gravitational settling and surface depletion. Other available air models use the source depletion method. The technical differences are described in a critique of the source depletion approach presented by Horst (1977). Wet deposition processes are approximated by applying a 'scavenging ratio'. 'Scavenging' is assumed to account for both vapor or air-water partitioning and removal of particulates by rainout. Finally, the DiDOT model is structured to provide loadings to land and water surfaces according to a grid network superimposed on the local landscape.

3.2. Land surface processes

The fate and transport of organic toxicants within and on soil systems arises from the interaction of pollutant-specific processes (e.g., degradation) and the hydrologic cycle (Bailey *et al.*, 1974). Transport is dominated by leaching, runoff, erosion and sediment transport. Because transport can occur in both water and sediment, pollutant partitioning between solution and particulate phases is necessary. The development of models to accommodate these processes has led to procedures designed for single land uses must be included because atmospheric loading to the land surface may result from widespread movement of pollutants rather than only localized interactions within a single land use.

The land component model chosen to be most compatible with the objectives in this study was EPA's NPS model. The NPS model was initially designed as an aid to the 208 water quality planning process and has been successfully applied in a number of mixed land-use watersheds (urban and rural). Two surface conditions are accommodated – impervious surfaces like streets and roofs, and pervious surfaces like lawns, parks, and rural areas. Impervious surfaces are simulated using standard urban stormwater analysis techniques and include surface accumulation of pollutants deposited from the atmosphere with subsequent washoff from precipitation-runoff events. Pervious land segments generate runoff based upon a continuous water balance that partitions incoming precipitation into soil moisture storage, evapotranspiration, infiltration, and surface runoff (rainfall or snowmelt). Erosion is calculated as a function of both rainfall intensity and runoff volumes.

The NPS model simulates pollutant movement to surface water by correlation to sediment transport. That is, the sediment (referred to as 'dust and dirt' in the case of impervious surfaces) predicted by the NPS model is assumed to transport a given amount of pollutant based upon the amount accumulated on the land surface and the extent to which the pollutant sorbs to sediments. This approach can lead to substantial errors if one attempts to predict the movement of very weakly sorbed organics. For this reason the choice of the NPS model may limit the immediate application of ALWAS to those pollutants that tend to be hydrophobic in nature. The NPS model is structured to accommodate loadings from the atmosphere such as those generated by the DiDOT model. More detailed discussion of the NPS model is found in the work of Donigian and Crawford (1976).

3.3. Surface water processes

Given appropriate organic toxicant loadings to surface water, (e.g. from the NPS or DiDOT models) the problem of estimating the fate and transport within selected aquatic environments remains. Smith *et al.* (1977) developed a steady-state, compartmentalized, process model to evaluate the interactions among various aquatic fate and transport phenomena. Later Burns and Cline (1982) expanded this work and developed a comprehensive organic chemical fate model based upon second-order kinetic descriptions of fate processes operating at steady-state within compartmentalized environments – the EXAMS model. EXAMS is an acceptable component for ALWAS with one notable

exception – calculations are steady-state but the loads generated from the other model components are time-varying. This limitation was circumvented by development of a time-varying version of EXAMS for use in ALWAS. The ALWAS 'EXAMS' permits loadings from streamflow, runoff (NPS model), direct atmospheric input (DiDOT model), and any other point source effluent (to either water column or benthic compartments) or groundwater input.

3.4. Water Treatment Processes

Exposure to toxicants in drinking water drawn from surface water sources usually occurs only after such water has been subjected to various treatment operations. Potable water treatment may vary from simple sedimentation/chlorination to chemical coagulation and activated carbon adsorption. Hedden (1980) developed the WTP model to analyze various combinations of possible treatments schemes for their ability to reduce toxicant concentrations prior to distribution through a water supply system. Unit process theory was applied to both dissolved and sorbed pollutants as they are subjected to a specified process train. The WTP model is not currently formally incorporated in ALWAS, but is included in the example application to follow.

4. Input Data Needs

Data required to execute a multimedia analysis are by no means trivial but are generally available from various inventories and national data centers. Hedden (1981) has described such data needs in detail and additional specific information is noted in the ALWAS documentation by Tucker *et al.* (1981). Input data are of four general types: (1) environmental loading rates for sources not explicitly simulated; (2) hydrologic and meteorologic time series; (3) specific media properties, and (4) pollutant fate equilibrium and rate constants.

A major objective of multimedia modelling is to predict intermedia transfers of toxicants, but to do this, the initial release(s) into the environment must be known. Stack emissions, wastewater discharges, and other point sources must be inventoried, located, and quantified. The hydrologic and meteorologic time series combine to determine many of the transport and dispersion processes. Such site-specific data (wind speed and direction, precipitation, streamflow, etc.) are usually collected continuously as part of weather station or stream flow recordings operated by the U.S. National Weather Service and U.S. Geological Survey. Similarly, other site-specific media properties (e.g., drainage areas, stack heights, land use, surface water depths) required to define the model boundary conditions and physical parameters are easily obtained from U.S. Dept. of Agriculture soils maps, topographic maps, planning agency records, and other generally available sources. Finally, pollutant-specific equilibrium and kinetic rate transformation process constants must be measured or estimated using appropriate structure-activity relationships. In many cases protocols have already been developed for laboratory measurement of such data (Mill *et al.*, 1981). The component models in ALWAS have

been selected such that the pollutant-specific data can be readily determined by execution of these protocols or can be inferred from existing literature.

5. Application Example

The ALWAS model has yet to be tested for a given location although some of its individual components have been subjected to tests and have been operationally applied (e.g., NPS model). Tucker *et al.* (1981) have applied the model to a site for which the NPS had been previously calibrated. To illustrate the potential use of ALWAS as an aid in multimedia exposure assessment, results of Tucker's work were combined with simulation of potable water treatment plant processes. The resulting estimated organic chemical concentrations in finished drinking water can be compared to the assumed environmental loadings to evaluate the multimedia transport and fate of the selected chemical. A watershed located within Durham, NC, drained by Third Fork Creek was selected as the site for the demonstration. Benzo(*a*)pyrene (BaP) was selected as the toxic organic pollutant to be evaluated. These choices were made solely because of prior hydrologic studies in the area and the desired properties of the toxicant. The choice is not meant to imply a problem within the area or to suggest any specific future study be conducted in the area. Indeed, because simulation of a pond was desired, a hypothetical dam was assumed to be located at the watershed outlet. Thus the results of this assessment apply *only* to assumed or hypothetical conditions – *not* existing site conditions. Finally, the water treatment plant configuration shown in Figure 1 was adopted for use in the WTP model.

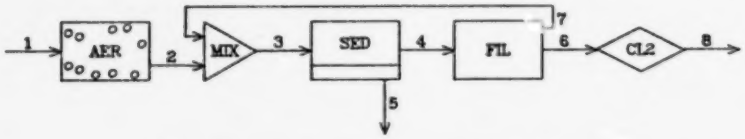

Fig. 1. Schematic for assumed water treatment plant.

5.1. Description of Study Area

The Third Fork Creek basin is located in the eastern section of the North Carolina Piedmont, a region of gently rolling hills. The stream flows in a southerly direction and is tributary to the New Hope, Haw, and Cape Fear River systems. Upper Third Fork Creek, simulated in this study, drains an area of 433 hectares (1069 acres) located within the city limits of Durham, NC. The drainage basin is primarily composed of two shallow valleys with relatively minor flood plains along the lower reaches of the streams. Surface runoff generally follows natural drainage paths with a few man-made channels. No storm sewer system exists; storm runoff is removed through street gutters, small pipes, and culverts under roads.

The region experiences a moderate climate without distinct wet and dry seasons. Summer storms tend to be brief, high-intensity thunderstorms and convective showers. Winter and spring storms are of longer duration and lower intensity and are caused by migratory low-pressure weather systems. Mean annual precipitation is approximately 1200 mm (47 inches) of which less than 25 mm (1 inch water equivalent) occurs as snowfall with little significant accumulation.

The Third Fork Creek basin, as shown in Figure 2, represents a typical urbanized area in the Piedmont region of the Southeastern United States. The basin encompasses a variety of land uses, including:

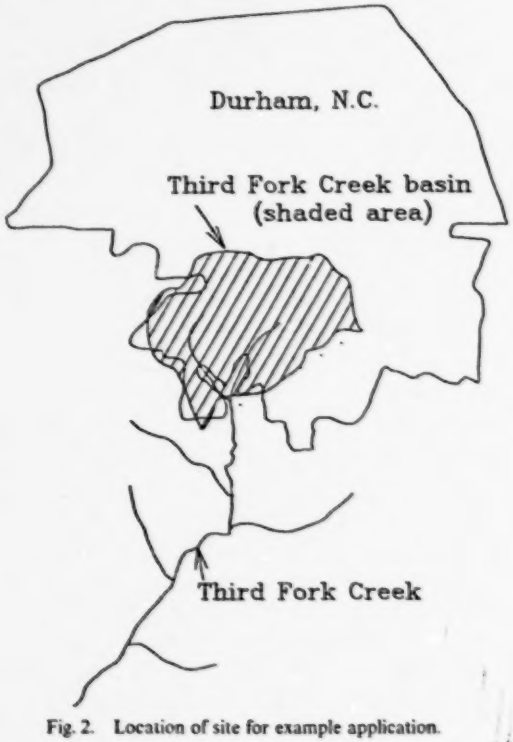

Fig. 2. Location of site for example application.

- high- and low-density housing units of varying quality,
- undeveloped land,
- shopping centers,
- a portion of the central business district,
- churches, schools, and small shops and stores,
- an urban redevelopment section,
- a tobacco processing plant,
- a section of expressway,

- a cemetery,
- slums,
- railroad yard, and
- a flood plain utilized mainly as a city park.

Air pollution emissions from areas other than the city of Durham will affect the study area. The DiDOT model can process emissions from nearby cities and emissions from the surrounding countryside up to four area source grids from the study area. These data requirements made it necessary to inventory emissions for all of Durham County and for the city of Raleigh.

The NPS model had been previously applied to this site permitting direct use of model parameters (e.g. erosion factors) obtained therein (Donigian and Crawford, 1976).

5.2. Development of input data

All sources and their ultimate BaP loadings must be estimated for the assumed pond. First the atmospheric loadings were input to the DiDOT model. Atmospheric loadings were located by developing a grid overlay for the watershed area and assigning appropriate point or area source strengths within each grid. Three atmospheric point sources for BaP in Durham were identified from data obtained from the North Carolina Department of Natural Resources. A total suspended particulate (TSP) emission rate was available for these sources, but a BaP emission rate was not. All the point sources were residual oil burning boilers. BaP emissions from these boilers were estimated by finding the ratio of nationwide BaP emissions (Nagda, 1979) to nationwide hydrocarbons (HC) emissions (NEDS, 1975) from residual oil burning boilers and multiplying the ratio by the point source HC emission rate. The estimated point source characteristics are given in Table I.

TABLE I

Point source characteristics

Source name	Emission rates TSP	(g/s) BaP[a] (estimated)	Stack height (m)	Stack diam (m)	Exit vel (m/s)	Stack temp (K)	Vol flux ($m^3 s^{-1}$)
Tobacco Processing A	1.07	1.46×10^{-4}	50.0	2.0		461.0	10.4
Durham County Hospital	0.14	3.64×10^{-5}	11.0	0.8	5.2	400.0	
Tobacco Processing B	1.73	5.10×10^{-4}	61.0	2.4		446.0	12.5

[a] BaP emissions were estimated via the ratio of national BaP emissions in residual oil boilers to the ratio of national hydrocarbon emissions for the same class of sources. These emission rates are used for illustrative purposes in the analysis only, not as an accurate estimate of BaP emissions.

Area source BaP loadings were estimated for transportation-related activities, rural-related activities (e.g., slash burning), and for the city area. All locally available information was used and the remaining emissions were estimated by using 'BaP ratios to

TSP' or 'a per population basis' with data taken from national values and adjusted to local conditions. Tucker *et al.* (1981) provide the detailed calculations. The results are given in Table II.

TABLE II

Summary of area source emissions used as input DiDOT

	Pollutant (tons yr^{-1})		BaP[a] (estimated)
	TSP	HC	
Rural	11136	1092	2.22×10^{-1}
Urban	6447	3443	7.14×10^{-1}
Total	17583	4535	9.36×10^{-1}
g/s-km^2 for INPUT to DiDOT			
Rural	0.42		8.38×10^{-6}
Urban	1.80		2.00×10^{-4}

[a] These emission rates are used for illustrative purposes in the analysis, not as accurate estimates of BaP emissions.

An additional constant environmental loading of BaP to the assumed pond was estimated. This loading was included to represent leaching from a disposal lagoon within the watershed and set equal to a constant, but low level of 6.0×10^{-7} kg $week^{-1}$ based upon the work by Falco *et al.* (1980).

The meteorological and hydrological data were obtained from the National Weather Service (NWS) recording station located at the Raleigh-Durham airport. Streamflow and watershed characteristics data for the site were those used by Donigian and Crawford (1976). The pond configuration assumed was that described by Burns and Cline (1982).

The selection of BaP as the toxicant for simulation was largely determined by two factors – it strongly sorbs to sediment (and hence is compatible with NPS transport algorithms) and the various rate constants required for EXAMS and the WTP model have been measured or estimated (Mabey *et al.*, 1981). Specific values, expressed in EXAMS-acceptable format, are given in Table III.

5.3. Results

The ALWAS as coupled to the WTP model and applied to the study site could be used to simulate many years of BaP concentrations in drinking water as a function of various media control strategies if desired. Indeed, one could argue that, for hydrologically driven systems, only after such a simulation period were analyzed could meaningful estimates of potential risk (or probabilities of exposure) be made. To illustrate the output results, however, only four months were simulated by using input data that began on June 1, 1972 and ended on September 30, 1972.

TABLE III

Transformation, physical, and transport data for Benzo(*a*)Pyrene

Property or process	Value
Photolysis data:	
Reaction quantum yield, 0, 313 nm	8.9×10^{-4}
Direct photolysis rate constant, k_p (hr^{-1}) *winter*, at 40° latitude midday	0.58
Oxidation constant at 25 °C:	
For $^1O_2$ (singlet oxygen), k_{OX} ($M^{-1}\,hr^{-1}$)	5×10^8
For RO_2 (peroxy radical), k_{OX} ($M^{-1}\,hr^{-1}$)	2×10^4
Hydrolysis rate constants:	
For base-promoted process, k_B ($M^{-1}\,hr^{-1}$)	0
For acid-promoted process, k_A ($M^{-1}\,hr^{-1}$)	0
For neutral process, k_N (hr^{-1})	0
Biotransformation rate constant:	
For bacterial transformation in water, k_b ($M^{-1}\,hr^{-1}$)	3×10^{-12}
Partition constants:	
Water solubility, S_w (ppm)	3.8×10^{-3} (25 °C)
Octanol-water, K_{ow} (unitless)	5.15×10^6
Sediment-water, K_{oc} (unitless)	5.5×10^6
Microorganisms-water, K_B ([μg/g] [mg/l]$^{-1}$]	1.5×10^5
Volatilization constants:	
Henry's constant, H_c (atm m^3 mol^{-1})	4.9×10^{-7}
Vapor pressure, P_v (torr)	5.6×10^{-9} (25 °C)

The modelling results can be examined at several steps in the overall calculation sequence. Because exposure via drinking water was the major interest here it seems reasonable to examine the total (dissolved and particulate) BaP concentrations in finished drinking water. Evaluation of these estimates in light of the source loading data and subsequent intermedia transfers should indicate the relative importance of alternative pathways and media control strategies.

The results of the four-month BaP simulation are shown in Figure 3. Note that the loadings to the unfinished water supply (the assumed pond) are given for each source. Loadings are expressed as total mass (both dissolved and sorbed) because BaP is highly sorbed and because the NPS model simulates loads as correlated to sediment. The lagoon loading is the only direct input – all others originate as direct loadings to the atmosphere and are subsequently transported to the pond via atmospheric processes (dryfall or wetfall) or via runoff from land surfaces. In the runoff case the dryfall and wetfall are first deposited on the land surfaces and subsequently transported to the pond in rainfall-runoff. The areal loading rate from wetfall/dryfall to land surfaces is not unlike that shown in the Figure 3 for the pond. The increased area over which such deposition occurs results in much greater mass accumulation and hence increased loading via runoff.

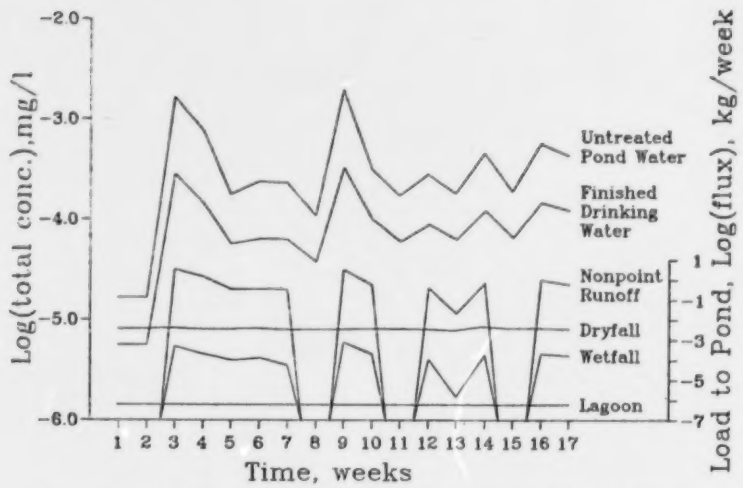

Fig. 3. Simulation results for multimedia exposure assessment of benzo(*a*)pyrone.

Figure 3 clearly shows that nonpoint or diffuse source loadings dominate the pond response. This follows from the substantially higher loadings (one to five orders of magnitude above all others for many weeks) in runoff and from the rapid hydraulic response of the assumed pond to runoff volumes. NPS model loads are reported here as weekly totals indicating runoff events during all but 5 of the total 17 week period. Indeed, 26 storms were simulated during the study period. The manifestation of random rainfall processes is clearly shown in the wide variation of simulated loadings.

Recall from prior discussion that BaP appearing as dryfall or wetfall and subsequent transport via runoff is subject to transformation processes (e.g. photolysis) during residence in air or soil. Similarly the loadings to the pond are subject to dispersion, advective transport, and reductions via transformations in the pond. The simulated net result is given as the total concentration identified in Figure 3 as unfinished drinking water. It is clear that total BaP concentrations in the pond respond directly to input loadings in general and nonpoint runoff loads in particular.

The assumed water treatment plant is a simple configuration that depends on sediment removal as the key process for BaP reduction. The total concentrations in treated water are also shown in Figure 3 and represent BaP removal efficiencies of from 67 to 84% over the simulation period.

The results demonstrated in Figure 3 illustrate only one of a number of possibilities one could investigate. For example, reductions in stack emissions could be compared with addition of more unit operations in water treatment, say carbon filtration, to examine cost-effective measures for reducing human exposure to BaP via drinking water. In a more general sense, through sensitivity testing, the array of loading rates and pollutant properties that combine to produce estimated drinking water toxicant concentrations

thought to be unacceptably high can be determined. Such information should prove helpful in evaluating new chemicals for their potential exposure levels.

6. Conclusions and Recommendations

The spatial and temporal variation of toxic chemical releases to the environment requires analyses of multimedia transport and fate if comprehensive exposure assessments are to be completed. Mathematical modelling offers a powerful tool toward that end. Existing, state-of-the-art, media-specific toxic organic transport and fate models for atmospheric, land surface, and surface water processes have been linked to demonstrate the technical feasibility of such an approach. Limited application and sensitivity testing of the modeling system has shown that the impact of various source loadings and control strategies on drinking water quality can be estimated.

The prototype model described here, ALWAS, is currently untested and warrants immediate investigation of its applicability to a wide range of problems. Specific modifications to accommodate pollutants that sorb only weakly to sediments appears to be a useful improvement. Because ALWAS was synthesized from independently developed model components, the overall model structure should be evaluated to determine more computationally efficient algorithms. The ALWAS should be applied to environments where monitoring has already occurred (or is contemplated) sufficient to investigate the model's predictive capability.

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